



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of Nobuo OI et al.

Serial No. : 09/972,879

Group Art Unit : 1772

Filed : October 10, 2001

Examiner : RHEE, JANE

For : COPOLYMER

\* \* \* \* \*

DECLARATION UNDER 37 C.F.R. §1.132

Honorable Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir:

I, Nobuo OI, a Japanese citizen residing at 4-3-13,  
Saginumadai, Narashino-shi, Chiba, Japan,  
declare:

That I am one of co-inventors of the above-identified  
application;

That I am familiar with the prosecution history of the  
above-identified application; and

That the following experiments were conducted by me or under  
my direct supervision.

**1. Purpose**

To know whether or not there is a significant difference  
between the copolymer in the claimed invention and the copolymer

disclosed in U.S. Patent 6,288,193 (Iseki et al.) cited in the previous office action mailed on December 30, 2002.

## 2. EXPERIMENT

### Experiment 1 (Claimed Invention)

Into a 400 ml-autoclave having an atmosphere replaced with argon were introduced 20.5 ml of vinylcyclohexane and 126 ml of dehydrated toluene. After heating to 30°C, ethylene was charged to 0.8 MPa. Further, 2.8 ml of a solution of methylalumoxane in toluene [MMAO produced by Tosoh-Akzo Corp., Al atom converted concentration=6 wt%] was charged and then a solution of 1.1 mg of isopropylidenebis(indenyl)zirconium dichloride in 1.1 ml of dehydrated toluene was charged. The reaction liquid was stirred for one hour and then poured into 500 ml of methanol, and a precipitated white solid was collected through filtration. The solid was washed with methanol and then dried under reduced pressure, to obtain 26.2 g of a polymer.

The polymer was subjected to purification by solvent fractionation. The polymer of 10.15g was subjected to Soxhlet extraction with chloroform for 10 hours, to obtain 9.75 g of a polymer as a component that was soluble in chloroform. The chloroform-soluble polymer had an intrinsic viscosity  $[\eta]$  of 0.63 dl/g and a vinylcyclohexane unit content in the polymer of 18 mol%.

The  $^{13}\text{C}$ -NMR spectrum of the polymer thus obtained is shown in Fig. 1A. It was confirmed by  $^{13}\text{C}$ -NMR signals in the vicinity of 34.1 ppm that the polymer had structure in which carbon atoms substituted with a cyclohexyl group were respectively separated by one methylene group of about ten percent based on the total

vinylcyclohexane units.

The above- and below-described measurements of the viscosity and vinylcyclohexane unit content and  $^{13}\text{C}$ -NMR spectrum analysis were carried out in the same manners as in Examples of the specification in the above-identified application.

#### Experiment 2 (Claimed Invention)

Into a 5000 ml-autoclave having an atmosphere replaced with argon were introduced 164 ml of vinylcyclohexane and 2780 ml of dehydrated toluene. After heating to  $50^{\circ}\text{C}$ , ethylene was charged to 0.4 MPa. Further, 47.1 ml of a toluene solution of methylaluminoxane [MMAO manufactured by Tosoh-Akzo Corp., concentration: 6 wt% in terms of Al atom] was charged, and subsequently a solution of 4.3 mg of isopropylidenebis(indenyl)zirconium dichloride dissolved in 8.7 ml of dehydrated toluene was charged. After the reaction mixture was stirred for one hour, the reaction mixture was poured into 2000 ml of methanol and a precipitated white solid was collected through filtration. The solid was washed with methanol and dried under reduced pressure to obtain a copolymer 76.0 g.

The copolymer had an intrinsic viscosity  $[\eta]$  of 1.88 dl/g and a vinylcyclohexane unit content in the copolymer of 5 mol%. The  $^{13}\text{C}$ -NMR spectrum of the obtained polymer is shown in Fig. 2A.

It was confirmed by  $^{13}\text{C}$ -NMR signals in the vicinity of 34.1 ppm that the polymer had structure in which carbon atoms substituted with a cyclohexyl group were respectively separated by one methylene group of about ten percent based on the total vinylcyclohexane units.

Further, the polymer was subjected to DSC measurement using

a differential scanning calorimeter (SSC-5200 manufactured by Seiko Instrument, Ltd.) under the same conditions as those in Examples of the specification in the above-described application.

As results, the melting point was 99.5°C, the calorie for fusion ( $\Delta H$ ) was 105 mJ/mg, and the crystallization temperature was 87.2°C.

### Experiment 3 (Comparative Example)

Into a 5000 ml-autoclave having an atmosphere replaced with argon were introduced 100 ml of vinylcyclohexane and 2000 ml of dehydrated hexane. After heating to 70°C, ethylene was charged to 1.2 MPa. Further, 1.5 ml of a solution of triisobutylaluminum in toluene [manufactured by Tosoh-Akzo Corp., concentration:1 mol/l] was charged and then a solution of 0.7 mg of dimethysilylene(tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride dissolved in 2 ml of dehydrated toluene and a solution of 4.1 mg of dimethylaniliniumtetrakis(pentafluorophenyl)borate dissolved in 2 ml of dehydrated toluene were charged. After the reaction mixture was stirred for one hour, the reaction mixture was poured into 2000 ml of methanol to collect a precipitated white solid. The solid was washed with methanol and dried under reduced pressure to obtain a polymer 48 g. The polymer had a vinylcyclohexane unit content of 5 mol%.

The  $^{13}\text{C}$ -NMR spectrum of the obtained polymer is shown in Fig. 3A.

It could not be confirmed that the viscous compound had structure in which carbon atoms substituted with a cyclohexyl group were respectively separated by one methylene group.

Further, the polymer was subjected to DSC measurement under the same conditions as in Experiment 2.

As results, the melting point was 99.4°C, the calorie for fusion ( $\Delta H$ ) was 104 mJ/mg, and the crystallization temperature was 82°C.

#### Experiment 4 (Comparative Example)

Into a 400 ml-autoclave having an atmosphere replaced with argon were introduced 27.4 ml of vinylcyclohexane and 166 ml of dehydrated toluene. After heating to 30°C, ethylene was charged to 0.8 MPa. Further, 3 ml of a solution of triisobutylaluminum in toluene [manufactured by Tosoh-Akzo Corp., concentration:1 mol/l] was charged and then a solution of 2.3 mg of dimethysilylene(tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride dissolved in 2.3 ml of dehydrated toluene and a solution of 9.2 mg of triphenylmethylenetetraakis(pentafluorophenyl)borate dissolved in 2 ml of dehydrated toluene were charged. Though the reaction mixture was stirred for one hour and then poured into 500 ml of methanol, no polymer was precipitated.

Therefore, the solvent containing methanol poured was removed from a mixture of the reaction mixture with methanol with an evaporator to obtain a copolymer as a viscous liquid having an intrinsic viscosity  $[\eta]$  of 0.24 dl/g and a vinylcyclohexane unit content in the copolymer of 18 mol%.

It could not be confirmed from  $^{13}\text{C}$ -NMR spectrum of the viscous liquid that the copolymer had a structure in which carbon atoms substituted with a cyclohexyl group were respectively separated by one methylene group.

### 3. CONSIDERATION

(1) It is apparent from Experiments 1 and 2 that the copolymer of the claimed invention had a structure in which carbon atoms substituted with a cyclohexyl group were respectively separated by one methylene group.

(2) However, it could not be confirmed that The copolymer prepared with the catalyst used in Iseki et al had a structure in which carbon atoms substituted with a cyclohexyl group were respectively separated by one methylene group.

(3) The crystallization temperature was significantly different between the copolymers of Experiments 2 and 3 though the vinylcyclohexane content was substantially the same each other.

(4) Accordingly, it is apparent that the structure and physical property of the claimed copolymer are significantly different from those of the copolymer disclosed in U.S. Patent 6,288,193 (Iseki et al.) .

That I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the above identified application or patent issued thereon.

Signed this 2nd day of June, 2005



---

Nobuo OI

Fig. 1A

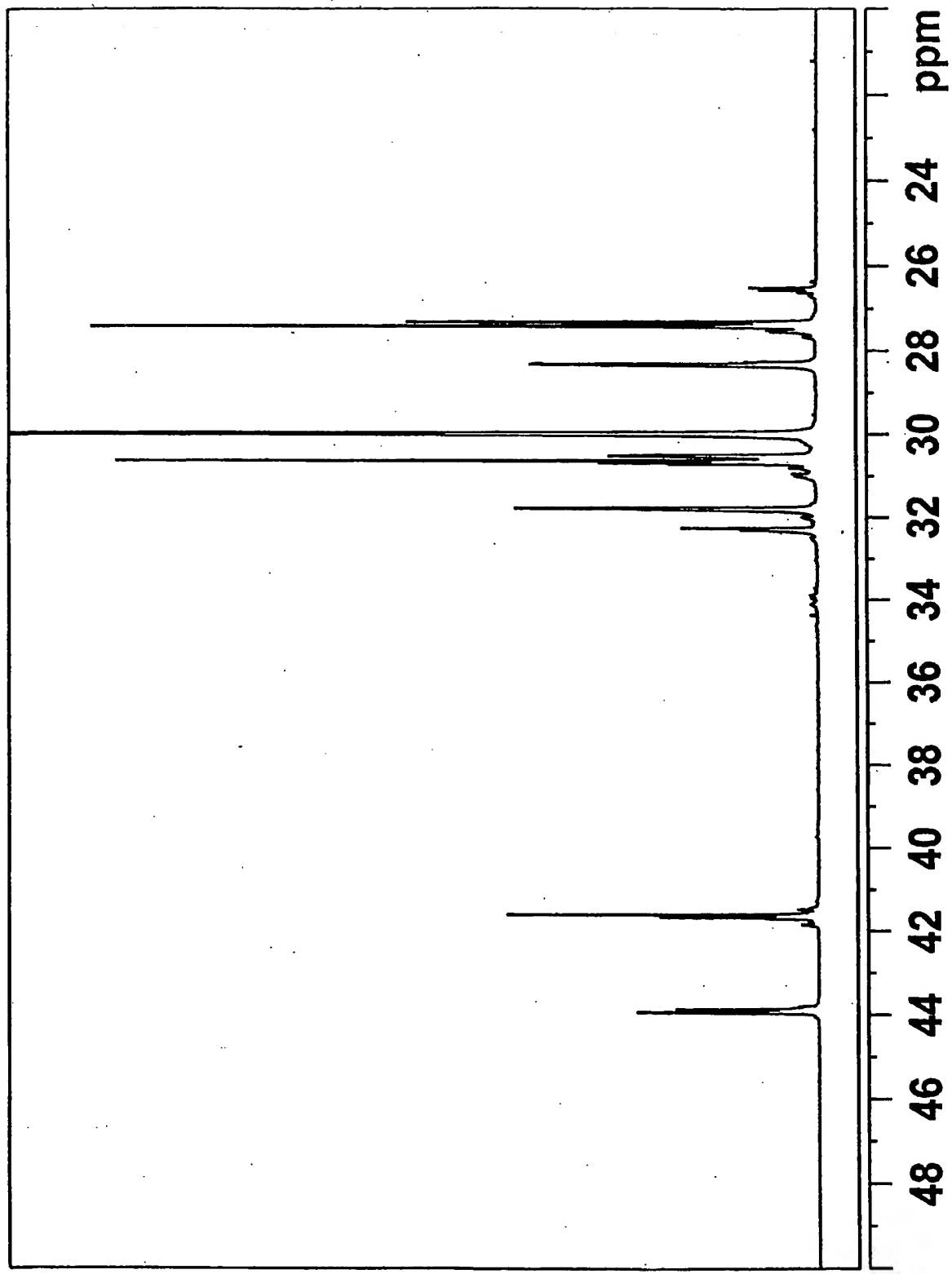




Fig. 2A

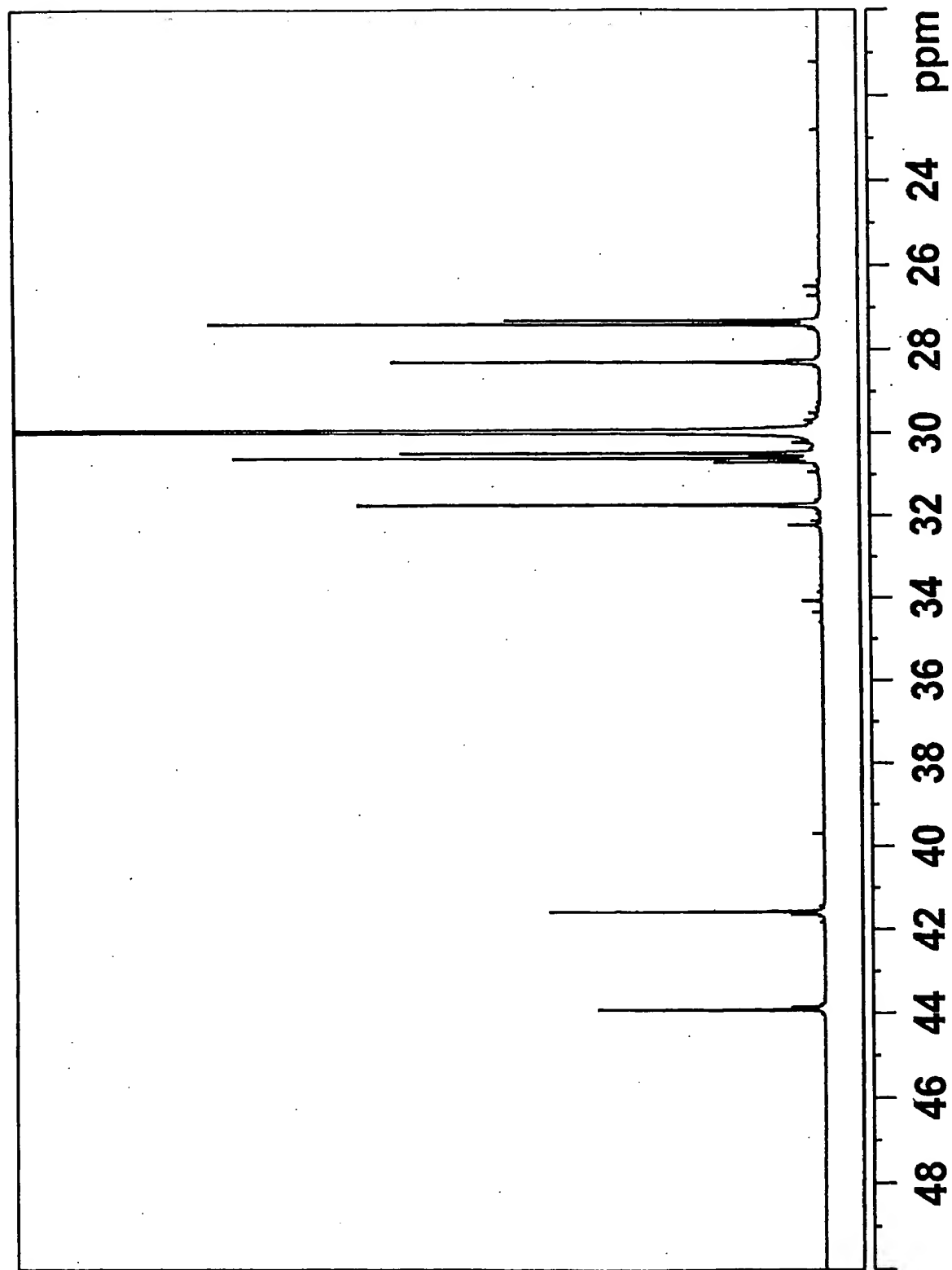


Fig. 3A

